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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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(21) International Application Number: PCT/BE95/00092 (22) International Filing Date: 9 October 1995 (09.10.95) (30) Priority Data: 9400911 7 October 1994 (07.10.94) BE (71) Applicant (for all designated States except US): ORFIT INDUSTRIES [BE/BE]; Vosveld 9a, B-2110 Wijnegem (BE). (72) Inventor; and (75) Inventor/Applicant (for US only): CUYPERS, Steven [BE/BE]; Swaenebeecklaan 39, B-2970 'sGravenwezel (BE). (74) Agents: VAN CUTSEM, Paul et al.; Bureau Vander Haeghen S.A., Rue Colonel Bourg 108A, B-1040 Bruxelles (BE).	(81) Designated States: AM, AU, BB, BG, BR, BY, CA, CN, CZ, EE, FI, GE, HU, IS, JP, KE, KG, KP, KR, KZ, LK, LR, LT, LV, MD, MG, MN, MW, MX, NO, NZ, PL, RO, RU, SG, SI, SK, TJ, TM, TT, UA, UG, US, UZ, VN, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG), ARIPO patent (KE, MW, SD, SZ, UG). Published <i>In English translation (filed in Dutch). Without international search report and to be republished upon receipt of that report.</i>	
(54) Title: METHOD FOR PRODUCING A THERMOPLASTIC SYNTHETIC MATERIAL AND IMMOBILIZATION ELEMENT PROVIDED THEREWITH (57) Abstract The present invention relates to a method for producing a thermoplastic material, wherein a thermoplastic base material is chosen which is admixed with at least one filler in the form of microspheres to obtain a foamed thermoplastic material having specific mechanical strength and flexibility, the obtained mixture being subjected to at least one further processing step, in particular extrusion. The invention also relates to a thermoplastic immobilization element for immobilization of a part of the body, which comprises a layer made from an intermediate produced by means of said method for (para)medical applications, orthopaedics, rehabilitation, for the purpose of supporting, replacing or fixating a part of the body, in radiotherapy and radiology and as an armouring protecting element in sport and the like.		

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METHOD FOR PRODUCING A THERMOPLASTIC
SYNTHETIC MATERIAL AND IMMOBILIZATION
ELEMENT PROVIDED THEREWITH

Field of the invention

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The present invention relates to a method for producing a thermoplastic material for immobilization and/or protection of a part of the body, wherein a thermoplastic base material is chosen which is admixed with at least one filler.

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Background of the invention

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Various applications are possible in this context, in particular for immobilization purposes in medical and paramedical applications, for protective purposes in sport and for use in the production of shoes.

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In the fields of application of orthopaedics, physical rehabilitation and sports medicine large amounts of plaster are still being used. Furthermore, in this context as in radiotherapy and radiology, a whole series of immobilization and fixation materials are employed such as thermoplastic plate materials and thermosetting bandaging materials.

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Prior art

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In the light of the abovementioned applications, the existing materials have the drawback of having a relatively high density. This high density

results in two significant troublesome problems with respect to the abovementioned applications: the materials have a relatively high weight which limits user comfort and which may even trouble the user, and
5 the therapeutic use of the materials used is diminished.

This may even lead to a medical problem, owing to the user being forced to greater exertion as a result of the greater weight.

The high density makes it impossible, in radio-
10 therapy applications, in particular those employing low-energy sources, to transmit radiation through such dense materials without this having an adverse effect on the skin of the patient. The materials in moulded form are placed on the skin of the patient and fixed to a
15 baseplate in order to put the patient in an immobile and reproducible position. These materials do not sufficiently transmit the radiation of, in particular, low-energy sources, as a result of which the radiation builds up immediately below the plastic material and can
20 thus damage the skin.

In radiology, the relatively high density of most existing materials or intermediates constitutes a problem in that they do not sufficiently transmit the types of radiation and thus produce interferences on the
25 image which may diminish the visibility of details on the image or may even cause distortion.

Object of the invention

30 It is an object of the invention to provide a solution to the abovementioned problems.

Exposition of the invention

35 To this end, according to the invention, there

is added, as the filler, a specific percentage of microspheres, said percentage being such that a foamed thermoplastic material is obtained which exhibits a specific mechanical strength and flexibility, said microspheres forming a foaming agent, and the obtained mixture of thermoplastic material and foaming agent is subjected to at least one further processing step. Thanks to the invention a foamed intermediate is obtained which is sufficiently strong and more user-friendly than existing intermediates, owing to which the invention primarily has a lower density and consequently a lower weight and better transmissivity for radiation sources which are used in radiotherapy and in radio-diagnostics and thus makes the invention further suitable for immobilization and/or protection of a part of the body.

In this context, according to an advantageous embodiment of the invention, the microspheres contain a lightweight gas which, when exposed to a specific temperature, causes the microspheres to expand within the mixture which is thus likewise subjected to an expansion process which makes it possible to effect a significant reduction in weight of the intermediate. In particular, said foaming agent is added in accordance with a percentage which is within the range from 0.1% to 10%, it thus being possible to achieve a reduction in density of the material of between 1 and 60%. Thus, in the course of the plastic being processed into an intermediate, the density of the plastic is lowered by a percentage of between 1% and 60%. Furthermore, the lighter an immobilization, fixation or protection material, the more pleasant it is for the user.

According to a generally advantageous embodiment of the invention, the mixing of said base material and said foaming agent is effected by

subjecting said mixture to a step involving a set temperature and a set residence time of the mixture in the extrusion means. In particular, during the extrusion step the temperature is set to such a value that an expansion process of the mixture is started under the influence thereof.

More in particular, said residence time of said mixture is set, during the extrusion step, within a time interval between a minimum value t_{min} , which is defined by a low time threshold, below which no expansion of said foaming agent takes place, and a maximum value t_{max} which is defined by a high time threshold, beyond which combustion of said mixture may take place.

According to a preferred embodiment of the invention, the expansion process of said mixture is tailored to the extrusion step and takes place completely within the latter, the expansion process being stopped, at the latest, when the extrusion step is stopped. If it isn't, the process continues during the processing of the intermediate, which leads to undesirable effects.

According to a further advantageous embodiment, said mixture is subjected to a multiple extrusion step, a multiple, in particular double, extrusion being carried out in the same step on said mixture. Thus better dispersion is obtained of the two components of the mixture, base material and foaming agent. Moreover, very accurate control of the processing temperature is thus accomplished.

This ensures, not least thanks to accurate control of the further extrusion parameters, in particular residence time, that the expansion process can be adequately controlled in order to arrive at a serviceable end product, owing to the foaming agent being well dispersed in the matrix base material. In

particular, in this case the base material chosen is a low-temperature thermoplastic which exhibits a processing temperature which is less than 100°C, preferably between 40°C and 90°C, more preferably between 50°C and 80°C. Alternatively it is also possible, in this context, for the base material chosen to be a high-temperature thermoplastic which exhibits a processing temperature which is greater than 100°C, preferably between 120°C and 180°C.

This is further assisted by the foaming agent being added in the form of a powder, in particular in a percentage within a range between 0.5 and 5%, preferably between 1 and 4%.

According to a yet further advantageous embodiment of the invention, said mixture is subjected to a two-dimensional extrusion, said foaming agent in particular being blended into resin granules or a so-called masterbatch, and said foaming agent is added in the form of the said resin granules. Thanks to this it was found that the foaming agent in the form of a masterbatch can be readily dispersed in the matrix base material. It was consequently possible to make the various base materials into a plate having a relatively smaller thickness, for example approximately 3 mm, while maintaining its advantageous properties according to the invention. Further, the microspheres have expanded uniformly in the polymer base material matrix, with a considerable reduction in density and likewise with a uniform distribution therein. In the process, foaming agent is added in a favoured manner at a percentage in the range between 2 and 6%, preferably between 3 and 5%, more preferably approximately 4%.

Hard foam materials based on thermosetting polyurethane do exist. This type of foam must, in order to provide sufficient mechanical strength, be used in a

relatively thick layer which makes it inappropriate for applications in radiotherapy and in radiology. Moreover, this type of foam must be prepared by the user him/herself at the time of application, on the basis of a 2-pack product which produces a chemical reaction. This process considerably reduces the user-friendliness of this type of foam. Moreover, it may also give rise to environmental problems.

In the case of hard foams based on poly(vinyl alcohol) the same problem of adequate mechanical strength arises, as a result of which it cannot be used, particularly in radiotherapy and radiological applications, as an immobilization or fixation material. Owing to the great layer thickness of the intermediates, these cannot be used on existing base fixation plates which are in circulation for these applications.

It is also possible, in accordance with alternative embodiments, for said mixture to be subjected to an injection-moulding process or to a pressure-moulding process, respectively.

In a supplementary manner it is further possible, after the completion of said at least one further processing step, in which an intermediate is obtained, for said intermediate to be subjected to a perforation.

According to a more specific embodiment, there is applied, to at least one of the two sides of the intermediate produced by means of said at least one further processing step, at least one further layer of plastic material having closed cells, to protect the surface against various external influences such as humidity and UV rays. In particular in this context, said at least one further layer is applied by means of a coextrusion step and it is further possible for each said further layer to have a different composition and/or structure.

According to a particularly advantageous embodiment, one of said further layers is applied as the outer layer (of the intermediate) which in this case is not foamed. This, surprisingly, proves successful in safeguarding the stiffness of the intermediate for a specific wall thickness while at the same time reducing the total weight of the intermediate with respect to an intermediate which consists entirely of a non-foamed material.

The present invention also relates to a thermoplastic immobilization element for the immobilization of a part of the body, which is remarkable in that it comprises a layer consisting of an intermediate which has been produced by means of a method as expounded hereinabove. Further characteristics of the thermoplastic immobilization element according to the invention are defined in the subordinate Claims 25 to 32 inclusive.

The invention further also relates to the use of the abovementioned thermoplastic immobilization element as a protective layer of a part of the body, in particular a limb, which is subjected to repeated frequent and accelerated movements, and of footwear, in particular a shoe having a heel and a tip, a thermoplastic immobilization element therein being provided as a stiffener thereof. The known materials which are presently used in the manufacture of shoes to stiffen the tips and the heels are relatively heavy, compared with the invention. Particularly in the case of sports shoes it is important that the weight remain as low as possible. The lower the weight of the component, the more efficient it is for the user.

The invention additionally also relates to an appliance whose characteristics are defined in the subordinate Claims 35 and 36.

Description

A few tests are described here. All tests have so far been done on extrusion machines.

5 In the first instance, a test was carried out on a laboratory extruder for making narrow ribbons. A first test was carried out employing, as the base material, polypropylene having a melting point higher than 100°C. (Use in so-called high-temperature thermoplastics, primarily in the orthopaedics sector;
10 less energy required for the activation of such materials than in the case of low-temperature thermoplastics, i.e. melting temperature below 100°C). The added foaming agent had the form of a powder. A first subtest was carried out with the addition of
15 almost 3% of foaming agent and a second subtest with the addition of almost 6% of foaming agent. These experiments produced the following observations:

- the combination of polypropylene and the foaming agent can be processed on conventional
20 extrusion machines, and the result according to the invention can consequently be achieved by means of ordinary classical apparatus;

- the temperature at which the extrusion occurs is a very important parameter for this
25 process, because it is the initiator of the expansion process;

- the residence time of the product in the extruder is likewise very important, since said residence time also the temperature
30 absorption affects the foaming agent [sic]. It was noted that if the residence time is too short, the foaming agent does not expand and if the residence time is too long, combustion of the material occurs; and

35 - the percentage addition of the foaming agent

to the base material further also plays an important part.

Production according to the invention takes place by a specific percentage of microspheres being added to the base material. These microspheres include a lightweight gas which, under the influence of temperature, increases in volume. Owing to this increase in volume, the microspheres take up a larger percentage of the volume of the intermediate. Since the expanded microspheres have a lower weight than the base material to which they were added, the weight of the intermediate is reduced as a result, the volume of the intermediate remaining the same.

For sports applications, the weight of the materials used is of primordial importance. Particularly in those applications, where provision of therapeutic immobilization or protection matters, the invention has a considerable advantage with respect to existing materials, in view of its low weight.

Whilst already foamed materials employed for this purpose do exist, such as softer foams based on polyolefins and polyurethane, these foams are too soft to be capable of an immobilization or fixation function and are therefore used exclusively as a comfort-creating element. They are therefore not eligible for the intended applications where immobilization or fixation is required, in contrast to the invention.

After the extrusion tests, the intermediates obtained were tested by heating them in an oven and forming them in accordance with the conventional techniques which are used in the respective fields of application. This showed that it is necessary for the expansion process to take place in its entirety during the extrusion. If this does not happen, the process continues during processing of the intermediate, which

leads to undesirable effects.

The extruded samples were studied under an electron microscope, which showed that the foaming agent in powdered form had indeed expanded but that it had nevertheless not dispersed entirely satisfactorily in the matrix base material.

Therefore, advantageously, in the second instance tests were carried out on a laboratory twin-screw extrusion machine. By means of such a machine, better dispersion is obtained of the two components of the mixture of base material and foaming agent. Moreover, this machine provides very accurate control of the processing temperature. In this test, the base material used comprised two thermoplastics which have a processing temperature below 100°C, in particular polycaprolactone and a blend of polycaprolactone and thermoplastic polyurethane. In each case, 1% and 3% foaming agent was added in the form of a powder. The result of the tests was that, given accurate control of the extrusion parameters, in particular temperature and residence time, the expansion process can be adequately controlled in order to arrive at a serviceable end product, provided that the foaming agent is sufficiently dispersed in the matrix base material.

A third test was carried out, this time on a production machine, viz. an extruder having a flat female mould of a width of approximately 120 cm for the production of flat plates. Again, various base materials were used as the matrix, viz. low-density polyethylene, polycaprolactone, and a blend of polycaprolactone and thermoplastic polyurethane.

In this test, the foaming agent was added in the form of a so-called masterbatch, viz. resin granules which had already been blended with the foaming agent. In each case, approximately 4% of masterbatch were

added.

These tests showed that the foaming agent in the form of a masterbatch is readily dispersible in the matrix base material. Consequently it was possible to
5 make the various base materials into a plate having a thickness of approximately 3 mm. After the extrusion tests, the plate materials were studied under an electron microscope, which showed that the microspheres had expanded evenly in the polymer matrix.

10 Moreover it was observed, when the test plates were weighed, that the density had decreased by from 27 to 33%.

Advantageously, the plastic material according to the invention is also used in sports, where, in
15 particular, materials to protect human limbs and for protecting the legs of horses in equestrianism are required. In the shoe industry likewise the plastic materials according to the invention are used for making stiffeners in a shoe.

20 The uses of the above-described invention are numerous, in particular for the use of a foamed thermoplastic material as an immobilization or fixation material of a part of the human or animal body in medical applications; for the use of a foamed
25 thermoplastic material in orthopaedics and/or physical rehabilitation, said material serving for the purpose of supporting, replacing or fixating a part of the human body; and for the use of a foamed thermoplastic material in radio-therapy and radiology, where a part of the body
30 is immobilized and positioned during the therapeutic treatment or the diagnostic investigation.

It would also be possible, according to a further advantageous application of the present invention, to combine the thermoplastic immobilization
35 element described here with that described in the

European Patent No. 0401883 of the same Applicant, this being incorporated by reference.

CLAIMS

1. Method for producing a thermoplastic material,
wherein a thermoplastic base material is chosen which is
5 admixed with at least one filler, characterized in that
there is added, as the filler, a specific percentage of
microspheres, said percentage being such that a foamed
thermoplastic material is obtained which exhibits a
specific mechanical strength and flexibility, said
10 microspheres forming a foaming agent, and in that the
obtained mixture of thermoplastic material as the base
material and foaming agent is subjected to at least one
further processing step.
2. Method according to Claim 1, characterized in
15 that the microspheres contain a lightweight gas which,
when exposed to a specific temperature, causes the
microspheres to expand within the mixture which is thus
likewise subjected to an expansion process.
3. Method according to any one of Claims 1 or 2,
20 characterized in that said foaming agent is added in
accordance with a percentage which is within the range
from 0.1% to 10%.
4. Method according to any one of Claims 1 to 3
inclusive, characterized in that the mixing of said base
25 material and said foaming agent is effected by
subjecting said mixture to an extrusion step with a set
temperature and a set residence time of the mixture in
the extrusion means.
5. Method according to the preceding claim,
30 characterized in that during the extrusion step the
temperature is set to such a value that an expansion
process of the mixture is started under the influence
thereof.
6. Method according to the preceding claim,
35 characterized in that the temperature is set within a

temperature range between 100°C and 200°C, preferably 160 and 180°C as the activation temperature for foams in the case of low-temperature thermoplastics.

7. Method according to any one of Claims 4 to 6 inclusive, characterized in that said residence time of said mixture is set, during the extrusion step, within a time interval between a minimum value t_{\min} , which is defined by a low time threshold, below which no expansion of said foaming agent takes place, and a maximum value t_{\max} which is defined by a high time threshold, beyond which combustion of said mixture may take place.

8. Method according to any one of Claims 4 to 7 inclusive, characterized in that the expansion process of said mixture is tailored to the extrusion step and takes place completely within the latter, the expansion process being stopped, at the latest, when the extrusion step is stopped.

9. Method according to any one of Claims 4 to 8 inclusive, characterized in that said mixture is subjected to a multiple extrusion step, a multiple, in particular double, extrusion being carried out in the same step on said mixture.

10. Method according to the preceding claim, characterized in that the base material chosen is a low-temperature thermoplastic which exhibits a processing or melting temperature which is less than 100°C, preferably between 40°C and 90°C, more preferably between 50°C and 80°C.

11. Method according to the preceding claim, characterized in that the base material chosen is a high-temperature thermoplastic which exhibits a processing or melting temperature which is greater than 100°C, preferably between 120°C and 180°C.

12. Method according to the preceding claim,

characterized in that foaming agent is added in the form of powder.

13. Method according to any one of Claims 10 to 12 inclusive, characterized in that said foaming agent is added in a percentage within a range between 0.5 and 5%, preferably between 1 and 4%.

14. Method according to any one of Claims 4 to 13 inclusive, characterized in that said mixture is subjected to a two-dimensional extrusion.

15. Method according to the preceding claim, characterized in that said foaming agent is first blended into resin granules (masterbatch) and that said foaming agent is further added in the form of said resin granules.

16. Method according to the preceding resin claim, characterized in that the foaming agent is added in a percentage in the range between 2 and 6%, preferably between 3 and 5%, more preferably approximately 4%.

17. Method according to any one of the preceding claims, characterized in that said mixture is subjected to an injection-moulding process.

18. Method according to any one of the preceding claims, characterized in that said mixture is subjected to a pressure-moulding process.

19. Method according to any one of the preceding claims, characterized in that after the completion of said at least one further processing step, in which an intermediate is obtained, said intermediate is subjected to a perforation.

20. Method according to any one of Claims 4 to 19 inclusive, characterized in that there is applied, to at least one of the two sides of the intermediate produced by means of said at least one further processing step, at least one further layer of plastic material having closed cells.

21. Method according to the preceding claim, characterized in that said at least one further layer is applied by means of a coextrusion step.

22. Method according to any one of Claims 20 or 21, characterized in that each said further layer has a different composition and/or structure.

23. Method according to any one of Claims 20 to 22 inclusive, characterized in that one of said further layers is applied as the outer layer (of the intermediate) which in this case is not foamed.

24. Thermoplastic immobilization element for immobilization of a part of the body, characterized in that it comprises a layer made from an intermediate produced by means of a method according to any one of the preceding claims.

25. Thermoplastic immobilization element according to the preceding claim where this depends on Claim 10, characterized in that said base material contains at least one thermoplastic such as polycaprolactone.

26. Thermoplastic immobilization element according to the preceding claim, characterized in that said base material contains at least one further base material such as thermoplastic polyurethane which forms a blend with said base material.

27. Thermoplastic immobilization element according to Claim 25, characterized in that said base material contains at least one further base material such as poly(vinyl chloride) or 1,4-trans-polyisoprene, which forms a blend with said base material.

28. Thermoplastic immobilization element according to any one of Claims 24 to 27 inclusive, characterized in that it comprises at least one further layer produced according to any one of Claims 20 to 23 inclusive.

29. Thermoplastic immobilization element according to Claim 24 where this depends on Claim 11,

characterized in that said base material is formed by an ionomer or an ethylene vinyl acetate.

30. Thermoplastic immobilization element according to Claim 24 where this depends on Claim 11,
5 characterized in that said base material is formed by a polypropylene.

32. Thermoplastic immobilization element according to any one of Claims 24 to 31 inclusive, having the characteristics from Claims 1, or 2 or 3, respectively,
10 of EP-0401883.

33. Use of a thermoplastic immobilization element according to any one of Claims 24 to 32 inclusive as a protective layer of a part of the body, in particular a limb, which is subjected to repeated, frequent and/or
15 accelerated movements.

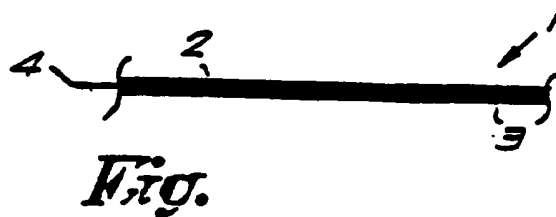
34. Use of a thermoplastic immobilization element according to any one of Claims 24 to 32 inclusive for footwear, in particular a shoe having a heel and a tip, the thermoplastic immobilization element therein being
20 provided as a stiffener thereof.

35. Appliance for implementing the method according to any one of Claims 9 to 13 inclusive, characterized in that it comprises a twin-screw extrusion machine.

36. Appliance for implementing the method according to any one of Claims 14 to 16 inclusive, characterized in that it comprises an extruder having a flat female mould having a certain width for fabricating flat
25 plates.

37. Method according to Claim 11, where this depends on Claim 5, characterized in that the activation temperature for the foaming in the case of a high-temperature thermoplastic is set within a temperature range which is between 180 and 250°C, preferably between
30 200 and 220°C.

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(54) Title: METHOD FOR PRODUCING A THERMOPLASTIC SYNTHETIC MATERIAL AND IMMOBILIZATION ELEMENT PROVIDED THEREWITH (57) Abstract The present invention relates to a method for producing a thermoplastic material, wherein a thermoplastic base material is chosen which is admixed with at least one filler in the form of microspheres to obtain a foamed thermoplastic material having specific mechanical strength and flexibility, the obtained mixture being subjected to at least one further processing step, in particular extrusion. The invention also relates to a thermoplastic immobilization element for immobilization of a part of the body, which comprises a layer made from an intermediate produced by means of said method for (para)medical applications, orthopaedics, rehabilitation, for the purpose of supporting, replacing or fixating a part of the body, in radiotherapy and radiology and as an armouring protecting element in sport and the like.		

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GA	Gabon				

INTERNATIONAL SEARCH REPORT

International Application No
PCT/BE 95/00092

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 C08J9/32 A61L15/07 A61F13/04

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 6 C08J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP,A,0 575 012 (SHELL) 22 December 1993	1-8, 11-14, 16,30, 36,37
Y	see page 3, line 19-21 see page 3; example 3; table 1 see claims 1,3,6,9	1-31, 33-37
Y	EP,B,0 393 003 (PONNET PAUL) 26 May 1993 see page 3, line 10-13 see figure see claims	1-31, 33-37

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents :

- *A* document defining the general state of the art which is not considered to be of particular relevance
- *E* earlier document but published on or after the international filing date
- *L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- *O* document referring to an oral disclosure, use, exhibition or other means
- *P* document published prior to the international filing date but later than the priority date claimed

T later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

X document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

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a document member of the same patent family

Date of the actual completion of the international search

20 February 1996

Date of mailing of the international search report

03.04.96

Name and mailing address of the ISA

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Oudot, R

INTERNATIONAL SEARCH REPORT

International Application No
PCT/DE 95/00092

C(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US,A,4 243 754 (SWAN JR.) 6 January 1981 see figures 2,7 see claims ---	1-31, 33-37
X	DATABASE WPI Section Ch, Week 8604 Derwent Publications Ltd., London, GB; Class A32, AN 86-023788 & JP,A,60 244 511 (ASAHI GLASS KK) , 4 December 1985 see abstract ---	17,18
A	GB,A,2 244 714 (SANYO CHEMICAL INDUSTRIES LTD.) 11 December 1991 see page 13-14, paragraph C see page 24; figures 1,3; table 1 see claims 1-30 ---	1-31, 33-37
X	EP,A,0 390 147 (THE B.F. GOODRICH COMPANY) 3 October 1990 see page 3, line 31-36 see examples 1,5 see claims ---	15
A	US,A,5 244 613 (HURLEY & AL.) 14 September 1993 see claims -----	1-31, 33-37

INTERNATIONAL SEARCH REPORT

International application No.

PCT/BE 95/ 00092

Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☒ Claims Nos.: 32 (not searched); 33-37 (partially)
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:
Claim 32 refers to an other document
Claims 33-37 the part referring to claim 32 has not been searched
3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest.
- ☐ No protest accompanied the payment of additional search fees.

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/BE 95/08092

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